IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

#17
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Applicant : S.R. Narayanan et al.
Serial No. : 09/489,514
Filed : January 21, 2000
Title : MEMBRANE-ELECTRODE ASSEMBLIES FOR DIRECT METHANOL FUEL CELLS

Art Unit : 1745
Examiner : Julian Mercado

Board of Patent Appeals & Interferences
Commissioner for Patents
Washington, D.C. 20231

BRIEF ON APPEAL

In response to the Final Office Action mailed on June 13, 2002 and pursuant to the Notice of Appeal filed on September 23, 2002, Applicants submit the following Brief on Appeal. A separate check for $320 is enclosed to cover the fee for this Brief as required by 37 C.F.R. §1.17(b).

(1) Real Party in Interest

The real party in interest is Jet Propulsion Laboratory of California Institute of Technology.

(2) Related Appeals and Interferences

None.

(3) Status of Claims

Claims 7-20 are pending and stand rejected.

CERTIFICATE OF MAILING BY FIRST CLASS MAIL

I hereby certify under 37 CFR §1.8(a) that this correspondence is being deposited with the United States Postal Service as first class mail with sufficient postage on the date indicated below and is addressed to the Commissioner for Patents, Washington, D.C. 20231.

December 3, 2002
Date of Deposit

Diane Wimperis
Signature

Diane Wimperis
Typed or Printed Name of Person Signing Certificate

(4) Status of Amendments

All amendments have been entered.

(5) Summary of Invention

The invention provides a procedure for preparing a water-based catalyst ink that incorporates hydrophobic components in the catalyst layer. The catalyst ink includes water, particles of a fluorocarbon polymer with a particle size of 1 to 4 microns, and a catalytic material. The catalyst ink can be prepared and applied at room temperature to the surface of a substrate, such as an electrolyte membrane or an electrode backing material, to form a catalyst coated electrode. In contrast to conventional TEFLON emulsion methods for introducing hydrophobic components into the catalyst layer, the present catalyst ink does not require a high temperature treatment step to activate the hydrophobic particles. Furthermore, the present process allows precise control of the particle size of the hydrophobic component of the catalyst ink. The catalyst ink, when applied to the surface of the substrate, achieves efficient removal of water produced by the electro-reduction of oxygen. The direct methanol fuel cells using the membrane electrode assemblies made according to the invention operate at low air flow rates and remove water at the cathode effectively with minimal use of evaporative processes.

(6) Issues

1. Whether the subject matter of claims 7-11, 13, 14, 18, and 20 would have been obvious over U.S. Patent No. 5,677,074 to Serpico et al. in view of Dupont Zonyl Fluoroadditives.

2. Whether the subject matter of claim 12 would have been obvious over U.S. Patent No. 5,677,074 to Serpico et al. in view of Dupont Zonyl MP 1100 Fluoroadditive Technical Information, and U.S. Patent No. 5,992,008 to Kindler.

3. Whether the subject matter of claims 15-17 and 19 would have been obvious over U.S. Patent No. 5,677,074 to Serpico et al. in view of the Dupont Zonyl MP1100 Fluoroadditive Technical Information, and U.S. Patent No. 4,524,114 to Samuels et al.

(7) Grouping of Claims

All claims stand or fall together.

(8) Argument

Obviousness cannot be established by "hindsight combination to produce the claimed invention. . .[I]t is the prior art itself, and the applicant's achievement, that must establish the obviousness of the combination." *In Re Dance*, 174 F.3d 1308 (Fed. Cir. 1999). In *In re Dembiczak*, 175 F.3d 994 (Fed Cir. 1999), the court held that a showing of a suggestion, teaching or motivation to combine prior teachings "must be clear and particular....Broad conclusory statements regarding the teaching of multiple references, standing alone, are not 'evidence.' " In addition, the courts have consistently held that merely because a structure or device is modifiable, does not make the modification obvious unless the prior art suggests the desirability of the modification. See e.g., *In re Laskowski*, 871 F.2d 115 (Fed. Cir., 1989) and *In re Gordon*, 733F.2d 900 (Fed. Cir. 1984).

Merely finding multiple references, each describing individual elements of a particular claim, is insufficient to support an obviousness rejection. It is well-settled that two or more references cannot be combined absent some motivation to make the combination in the first place. Moreover, this motivation cannot be the result of a hindsight-guided determination. Hindsight, however, is precisely the basis for the proposed combination here.

Claims 7-11, 13, 14, 18, and 20 are patentable over Serpico et al. (U.S. Patent No. 5,677,074) in view of the Dupont Zonyl reference. Serpico, the primary reference, discloses a catalyst layer that contains catalyzed carbon particles that are intimately mixed with a nonionic fluoropolymer (e.g., PTFE). Serpico teaches a broad range of particle sizes spanning four orders of magnitude, from 0.05 μm to 500 μm (col. 2, lines 42-43), and preferably from 50 μm to 500 μm (col. 2, line 58). Serpico, therefore, does not recognize the importance of controlling particle size within a narrow range generally, and certainly not within the specific range of 1-4 microns called for in the claims, for the purpose of making a catalyst ink. On the contrary, the implication in Serpico is that particles having a broad range of sizes are equally suitable. In view of this directive, there would be no reason for a person of ordinary skill to select particles having

a very narrow size distribution of the type described in the Dupont Zonyl reference. Indeed, to do so would involve ignoring what Serpico teaches. This is impermissible. Accordingly, the rejection of claims 7-11, 13-14, 18, and 20 cannot stand and must be withdrawn.

The Examiner asserts that both Serpico et al. and the Dupont Zonyl reference are disclosures which are pertinent to the use of catalyst inks and thus, the references are mutually relevant. This is, however, incorrect. The Serpico et al. reference teaches a catalyst layer that contains catalyzed carbon particles that are intimately mixed with a nonionic fluoropolymer (e.g., PTFE). The fluoropolymer performs the role of a binder for the catalyst particles (see, for example, col. 4, lines 40-43 and FIG. 2). Consistent with this purpose, Serpico teaches melting or otherwise causing the particles to flow, thereby coating the catalyst particles. This is not the case for the Zonyl particles either as described in the Zonyl product literature or in the applicants' process. The Zonyl product literature describes a very different function for these particles (i.e., as an additive in the preparation of printing inks and coatings for improving ink properties, such as gloss, rub resistance, surface smoothness, and slip). In the applicants' process, the Zonyl particles also are not performing the role of a binder. Rather, they cause hydrophobic disturbances within the catalyst layer. Consistent with this function, applicants apply the catalyst layer at room temperature. Therefore, it would not have been obvious for one skilled in the art to merely substitute the particles in the Serpico reference (which are used as binder) with those in the Zonyl reference (which are used as an additive in printing inks and coatings) in order to achieve the present invention. Accordingly, the claims would not have been obvious in view of the combination of cited references and the rejection should be withdrawn.

The tertiary references, Kindler (U.S. Patent No. 5,992,008) and Samuels et al. (U.S. Patent No. 4,524,114) have been cited to describe additional features found in dependent claims 12 and claims 15-17 and 19, respectively.

For example, the Kindler reference has been cited to show that a liquid copolymer of tetrafluoroethylene and perfluorovinylethersulfonic acid can be employed for enhancing ionic conduction within an electrode.

The Samuels et al. reference has been cited to show that the surface of an electrolyte membrane can be roughened using silicon carbide prior to catalyst deposition.

The tertiary references, however, do not remedy the deficiencies of the Serpico and Zonyl references and provide no clear and particular suggestion that would have motivated one of ordinary skill in the art to combine the teachings of Serpico et al. and the Dupont Zonyl reference to achieve the presently claimed process. Furthermore, since the Serpico and Zonyl references fail to describe the subject matter of the independent claims from which these claims depend, the rejection of the dependent claims must fail as well.

For these reasons, Applicants respectfully submit that the present claims would have been unobvious over Serpico et al. in combination with the secondary references. The Board of Patent Appeals and Interferences is respectfully requested to reverse all rejections.

(9) Conclusion

For the reasons advanced above, the Board should reverse the rejections of claims 7-11, 13, 14, 18, and 20.

The brief fee of $320 is enclosed. Please apply any other charges or credits to Deposit Account No. 06-1050.

Respectfully submitted,

Date: 12/2/02

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Appendix of Claims

7. A process for making a catalyst ink for a fuel cell, comprising mixing, at room temperature, components comprising water, particles of a fluorocarbon polymer with a particle size of 1 to 4 microns, and a catalytic material.

8. The process of claim 7, wherein the particles have a surface area of about 5 to about 10 m^2/g.

9. The process of claim 7, wherein the catalytic material comprises Pt.

10. The process of claim 7, wherein the fluorocarbon polymer is selected from the group consisting of polytetrafluoroethylene polymers and fluorinated ethylene-propylene polymers.

11. The process of claim 7, wherein the catalyst ink further comprises an ionomer.

12. The process of claim 11, wherein the ionomer comprises a liquid copolymer of tetrafluoroethylene and perfluorvinyletherosulfonic acid.

13. A process for making an electrode assembly for a fuel cell, comprising:

(a) providing a catalyst ink comprising water, particles of a fluorocarbon polymer with a particle size of 1 to 4 microns, and a catalytic material; and

(b) applying the catalyst ink at room temperature to at least one side of a substrate.

14. The process of claim 13, wherein the substrate is a membrane.

15. The process of claim 14, further comprising roughening the side of the membrane prior to applying the catalyst ink.

16. The process of claim 15, wherein the side of the membrane is roughened by contacting the membrane with an abrasive selected from the group consisting of silicon nitride, boron nitride, silicon carbide, silica and boron carbide.

17. The process of claim 16, wherein the abrasive has a grit size of about 300 to about 400.

18. A process for making a membrane electrode assembly for a fuel cell, comprising:

(a) providing a catalyst ink comprising particles of a fluorocarbon polymer with a particle size of 1 to 4 microns, and a catalytic material;

(b) applying the catalyst ink at room temperature to at least one side of a membrane; and

(c) bonding the membrane to at least one electrode.

19. The process of claim 18, further comprising roughening the side of the membrane prior to applying the catalyst ink.

20. A fuel cell comprising a membrane electrode assembly, wherein the membrane electrode assembly is made by the process of:

a) providing a catalyst ink comprising particles of a fluorocarbon polymer with a particle size of 1 to 4 microns, and a catalytic material;

(b) applying the catalyst ink at room temperature to at least one side of a membrane; and

(c) bonding the membrane to at least one electrode.